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RADIATIONS FROM TWO RADIOACTIVE ISOTOPES OF GOLD†

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(*Received for publication March* 14, 1956

ABSTRACT. The angular correlation function of the 330kev-358 kev cascade in the decay of Au^{196} has been measured and is in accord with a 2-2-0 sequence of spins for the low lying states of Pt^{196}. The extent of anisotropy at 180° suggests the first emitted gamma transition to be a mixture of E2 in M1. The gamma radiation of the 185-day Au^{195} has been examined by scintillation counting methods and is found to consist of quanta at 31 kev and 99 kev in coincidence and the associated cross-over transition at 130 kev.

INTRODUCTION

Naturally occurring platinum was irradiated by deuterons of energy 15 Mev for 4.1 hours at an average beam current of 75 micro-amperes in the cyclotron at the University of Pittsburgh. The radioactive isotopes of gold so produced were chemically separated from iridium, mercury, and platinum. The gamma-ray spectrum, as measured in NaI(Tl) crystal and two weeks following the cessation of irradiation is shown in figure 1. The apparatus is described in a previous paper, Potnis *et al.* (1956). Photopeaks are in evidence at 65, 158, 350, and 425 kev. The 158 kev gamma ray was identified by its decay period as being emitted in the disintegration of Au^{199}, and the 350 kev and 425 kev quanta could similarly be assigned to Au^{196}. The x-ray energy of 65 kev is characteristic of the region of the noble metals. Calibration points for the spectrometers were obtained at quantum energies of 31.4, 87, 279, and 661 kev. The radiations of these energies were obtained from monoenergic gamma ray emitters such as Cd^{109}, Hg^{203}, and Cs^{137}. The 31.4 kev x-rays were those of Ba^{137} which are emitted following conversion of the 661 kev gamma ray.

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Au^{196} decays to Pt^{196} by orbital electron capture and the subsequent emission of a 330 kev—358 kev gamma-ray cascade. The radionuclide also decays

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Assisted by the joint programme of the Office of Naval Research and the U.S. Atomic Energy Commission.

by negatron emission followed by the gamma ray at 425 kev. The angular correlation function of the gamma-ray cascade in Pt^{196} has been previously measured by Steffen (1951, 1953). His results indicated a spin assignment of 2 to the first and second excited states of Pt^{196} and a scheme of 2-2-0 with the first emitted gamma-ray transition, a mixture of 95 percent E2 in M1. Sources in the form of a dilute solution of $AuCl_3$ as well as in the form of solid $AuCl_3$ imbedded in gold gave within the statistical errors, the same correlation function.

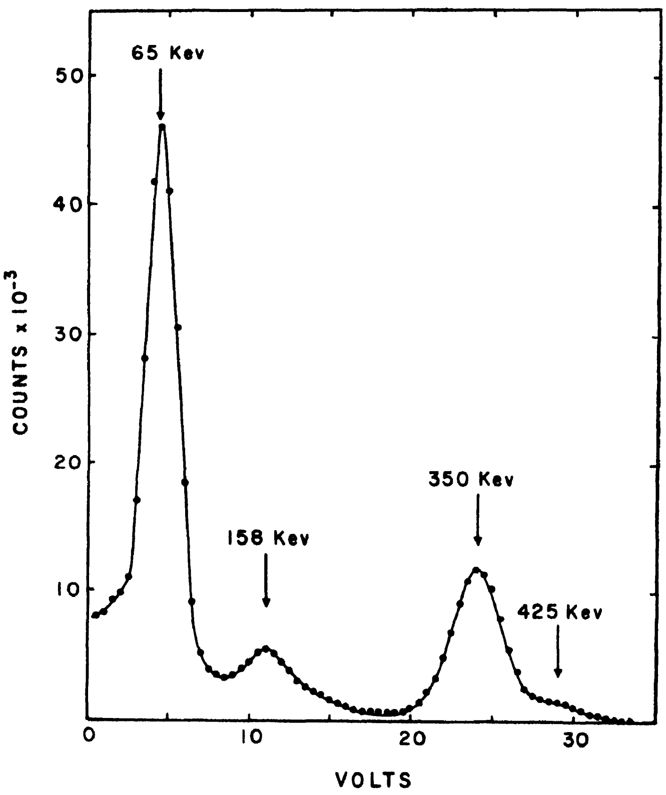

Fig. 1. Energy spectrum of gamma rays from isotopes of gold.

The correlation function of the 330 kev—358 kev cascade has been measured in the present investigation. Pulse height selection was employed in either channel, each being set at the photopeak appearing at ~ 350 kev shown in figure 1 and opened to a width of four volts. The resolving time of the coincidence circuit was 0.2 microsecond. The source was in the form of metallic gold contained in a carbon cylinder. The distance of the source from the face of either crystal was 13 cm, and the half-angle of the detecting system was 7.5 degrees as measured by the coincidence rate of the annihilation radiation of Na^{22}. An initial test of the proper function of the apparatus was carried out by measuring the anisotropy of the gamma-gamma coincidences of $Co^{60} \xrightarrow{\beta} Ni^{60}$. Measurements were performed at five different angles, the moving counter being placed at intervals of 22.5 degrees between the angles of 90 and 180 degrees with the axis of the fixed counter. Coincidences were accumulated at each angle for a period of five minutes at a time. This range of settings was traversed repeatedly so that any decay correction was eliminated. Approximately 10,000 counts were obtained at each angle.

The results of the measurements are presented in figure 2 where the observed

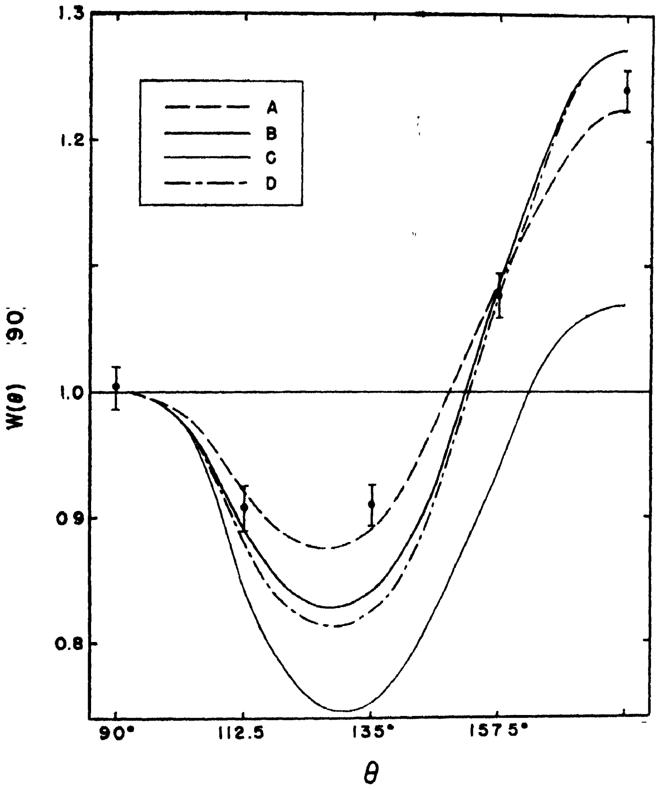

Fig. 2. Angular correlation function of the 330 kev—358 kev cascade in the de-excitation of Pt^{196}. Observed points are shown with statistical errors. Curve A—Least square fit of the data. Curve B—Curve A corrected for angular resolution of the detectors. Curve C—Theoretical correlation function for decay scheme 2(E2); 2(E2); 0. Curve D—Expected for a decay scheme 2(E2, M1); 2(E2); 0.

points are shown together with their respective statistical errors. A "least square" fit of the data yielded the function

$$W(\theta) = 1-0.66 \cos^2 \theta+0.88 \cos^4 \theta$$

where the probable errors of the coefficients of the terms in cos θ are about three percent. When the function is modified for the finite angular resolution of the apparatus, it becomes

$$W(\theta) = 1-0.90 \cos^2 \theta+1.17 \cos^4 \theta.$$

This latter curve is also plotted in figure 2 along with the theoretically expected distribution for a 2—2—0 spin sequence and both transitions pure electric quadrupole. The observed anisotropy at 180 degrees is 0.27 which is larger than would be expected for the pure cascade. A theoretical distribution function with the first emitted quantum a mixture of 96.7 per cent E2 in M1 is also plotted and agrees well with the observed function corrected for angular resolution. Thus is indicated the fact that the first transition occurs as a mixture with the above mentioned intensity ratio. The sign of the ratio of the matrix elements of the two types of transition was found to be positive, corresponding to a phase difference of 180°. Data available with regard to the internal conversion coefficients of the gamma rays are consistent with the condition that both transitions be electric quadrupole in character. The correlation observations of the present investigation are in essential agreement with those already obtained by Steffen (1951, 1953). Thus the spin assignments support the shell model predictions for an even-even nucleus like Pt^{196}.

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After a time of decay of about three months, the source previously employed in the study of Au^{196} was used to measure the radiations of Au^{195}.

Gold (195) is known to decay to excited states of Pt^{195} by orbital electron capture. The radiations emitted in this process have been examined in magnetic spectrometers and coincident Geiger counter arrangements, and several energy level schemes for Pt^{195}have been proposed. Steffen *et al.* (1949) found two non-coincident gamma rays with energies of 95 and 129 kev. De-Shalit *et al.* (1952) reported 29 and 97 kev gamma rays in cascade and a cross-over transition at 126 kev. Gillon *et al.* (1954) have observed conversion lines corresponding to the gamma-ray energies of the cascade but did not detect any cross-over transition.

The pulse-height distribution of the gamma rays of Au^{195}, as measured in a scintillation spectrometer, is shown in figure 3. Photopeaks are in evidence at quantum energies of 32, 65, 99 and 130 kev. The 32 kev photopeak is actually a composite one formed by the 37 kev escape peak of the 65 kev x-rays of Pt and the 31 kev gamma ray. When the pulses of this peak were absorbed in copper, two slopes were obtained corresponding to energies of approximately 31 and 65 kev, showing the presence of photoelectric pulses of a 31 kev gamma ray as well as those of the escape peak of the x-rays. These data are shown in the absorption curves of figure 4. From the counting rates at zero absorber thickness, it is estimated that 14 per cent of the pulses in the peak arise from the 31 kev gamma ray itself. The pulses of the 131 kev peak were similarly absorbed as is also plotted in figure 4, and the slope of the curve suggests an energy of 130 kev. Thus is eliminated the possibility that this peak arose from the simultaneous detection of the 31 kev and 99 kev gamma rays which are in cascade. Thus, in addition to the x-rays of platinum, it has been shown that three gamma rays are present in the decay of Au^{195}, the cascade, and the associated cross-over

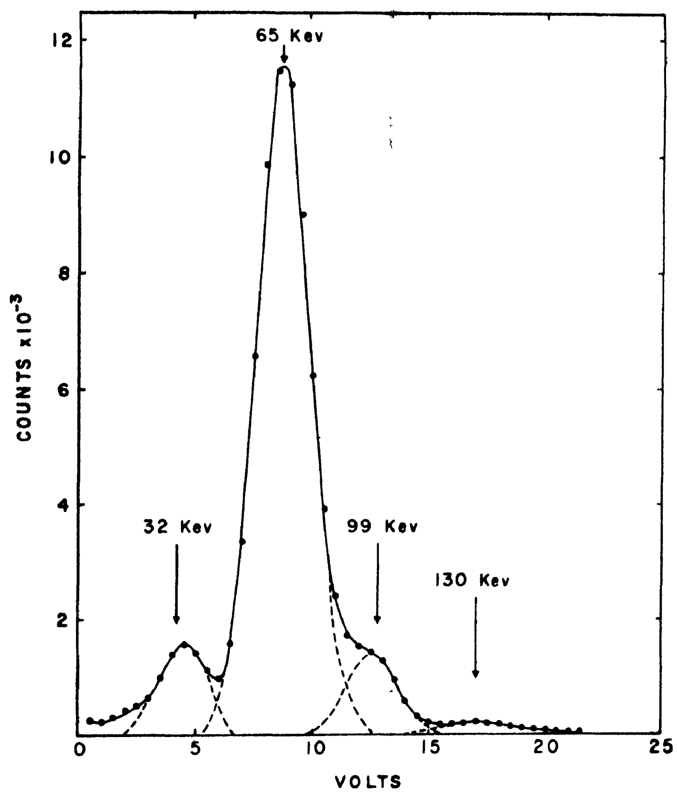

Fig. 3. Energy spectrum of gamma rays from Au^{195}.

transition. The 100 and 130 kev gamma rays are also observed in the proton and alpha-particle bombardment of natural Pt and assigned to Pt^{195} by Stelson and McGowan (1955). Gamma rays of energies 29, 98, 128, 210 and 240 kev have been observed in the electric excitation of Pt^{195} by Bernstein and Lewis (1955). The relative intensities of the unconverted quantum radiations can be estimated from the areas under the photopeaks of figure 2. They are 1, 12, and 2 in order of ascending energy. In making this estimate, corrections were applied for variation of the detection efficiency of the crystal with energy and the similar variation of the photopeak to Compton cross-section ratio.

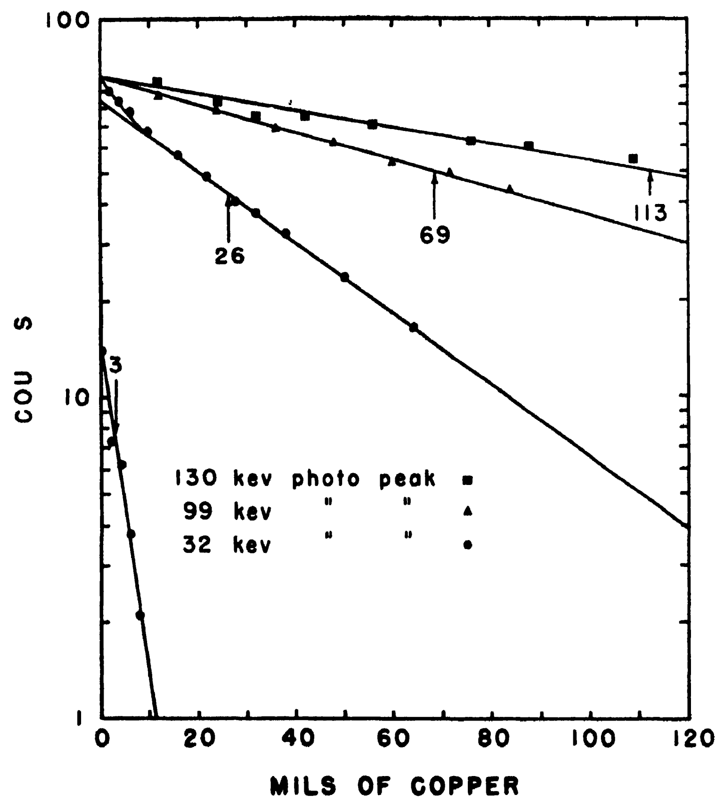

Fig. 4. Absorption of photopeaks of figure 3 in copper. Arrows indicate half-value thickness in mils of copper. Values 113, 69, 26, and 3 mils of copper correspond to gamma rays of 130, 99, 65, and 31 kev.

Coincidences between the gamma rays were measured, and the results are shown in figure 5. With one channel fixed at 99 kev., the data of figure 5A were obtained showing no detectable coincidences between the 99 kev gamma ray and any radiation of energy 130 kev. This shows that 130 kev gamma ray is a cross-over transition of the 31 and 99 kev gamma-ray cascade. With one channel fixed at the x-ray peak, the data of figure 5B were obtained, showing coincidences between the x-rays and the three gamma rays.

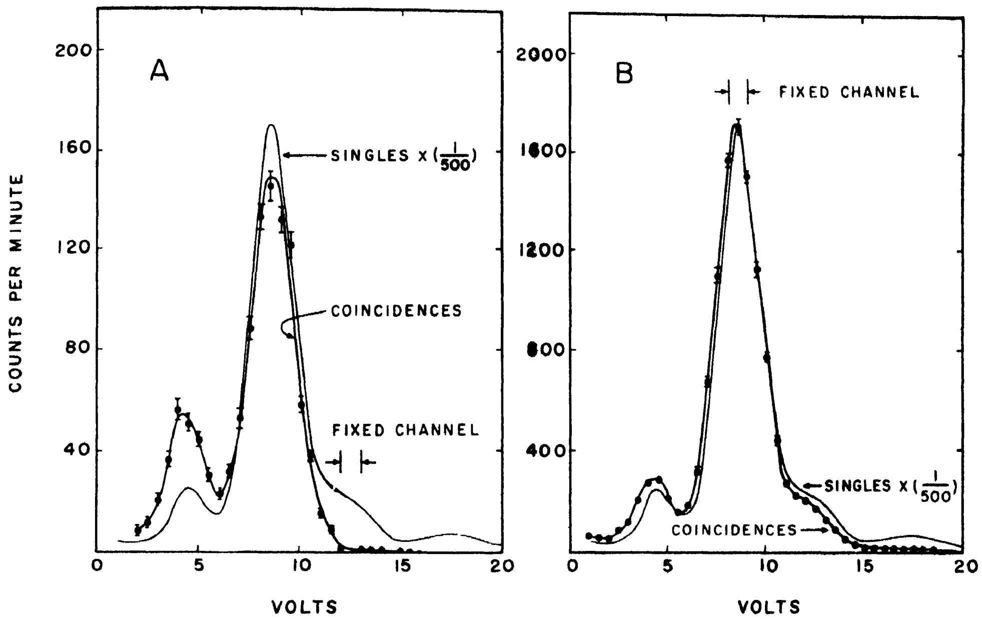

Fig. 5. (A) Gamma-gamma coincidences with 99 kev gamma ray.
(B) Gamma-gamma coincidences with 65 kev x-rays.

The total conversion coefficients of the various gamma rays have been previously measured, and when they are combined with the relative intensities of the presently measured unconverted quantum radiations, the transition probabilities of Table I are obtained. From a consideration of these transition intensities, it can be concluded that the 31 kev gamma ray is the first emitted of the cascade.

TABLE I

Energy, kev	Unconverted quantum intensities	α_T	Reference	Relative transition probability
31	1	7.3	De-Shalit	8.3
99	12	9	De-Shalit	120
		3.15	Steffen	49.8
130	2	1.28	Steffen	4.8

Depending upon which of the two values of the conversion coefficient of the 99 kev gamma ray is employed, the percent of capture transitions terminating at the

130 kev level is calculated to be 11 or 23. Previously reported values are 10 and 35 percent.

The decay scheme of Au^{195} is shown in figure 6. The ground state spin 1/2 of Pt^{195} has been measured by Jaeckel and Kopfermann (1936) in agreement with

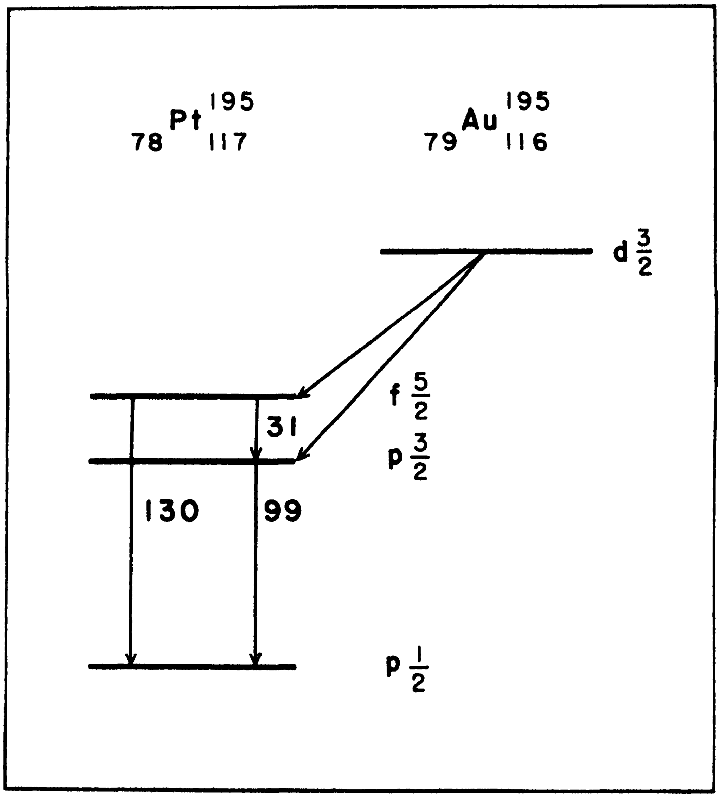

Fig. 6. Decay scheme for Au^{195}.

the orbital $p_{\frac{1}{2}}$ as indicated by the shell model. The data of Cork *et al* (1954) on the conversion in K- and L- shells for 31 and 99 kev gamma rays favour an assignment of M1 for both the gamma rays. The shell model indications of orbitals $p_{3/2}$ and $f_{5/2}$ to 99 and 130 kev levels agree with these assignments. With these assignments the 130 kev gamma-ray becomes E2 in nature. The measured conversion coefficients for this gamma ray (referred to in Table I) are not totally inconsistent with this classification. A spin of $d_{3/2}$ could be assigned to the ground state of Au^{195} on the shell model considerations.

The foregoing results are to be compared with those obtained by Cork *et al* (1954) and Potnis *et al* (1956) who have investigated energy levels in Pt^{195} by way of the decay of Pt^{195m}. In the work of Cork *et al*, the cross-over transition was not reported, whereas it was observed by the second group of authors. Their work is confirmed by the present investigation.

ACKNOWLEDGMENTS

The author wishes to acknowledge the keen interest of Dr. W. F. G. Swann, Director of the Bartol Research Foundation, and Dr. C. E. Mandeville for helpful suggestions, and to thank Dr. W. B. Keighton for chemical purification, to Mrs. Patterson and Mrs. Brandt for making drawings, and to Mrs. Rodgers for typing the manuscript. The author's visit to the United States was made possible by a scholarship granted by Bartol Research Foundation.

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